A Closed Loop Process for the End-of-Life Electric Vehicle Li-ion Batteries

Yan Wang (P.I.), Zhangfeng Zheng, Mengyuan Chen

Organization: Worcester Polytechnic Institute

2018 DOE Annual Merit Review

Date: June 18~21, 2018

Project ID #: bat293

Overview

Timeline

- Project start date: Feb 2, 2016
- Project end date: Feb 15, 2018, NCE to May 15, 2018
- Percent complete: 100%

Barriers

- Barriers addressed
  - Cost
  - Performance

Budget

- Total project funding: $1,024,740
  - DOE share: $512,370
  - Contractor share: $512,370
- Funding received in FY 2017: $361,137
- Funding for FY 2018: $169,510

Partners

- Interactions/ collaborations: A123 Systems, Battery Resourcers, Argonne National Laboratory, General Motors, Ford, FCA, SNT
- Project lead: WPI

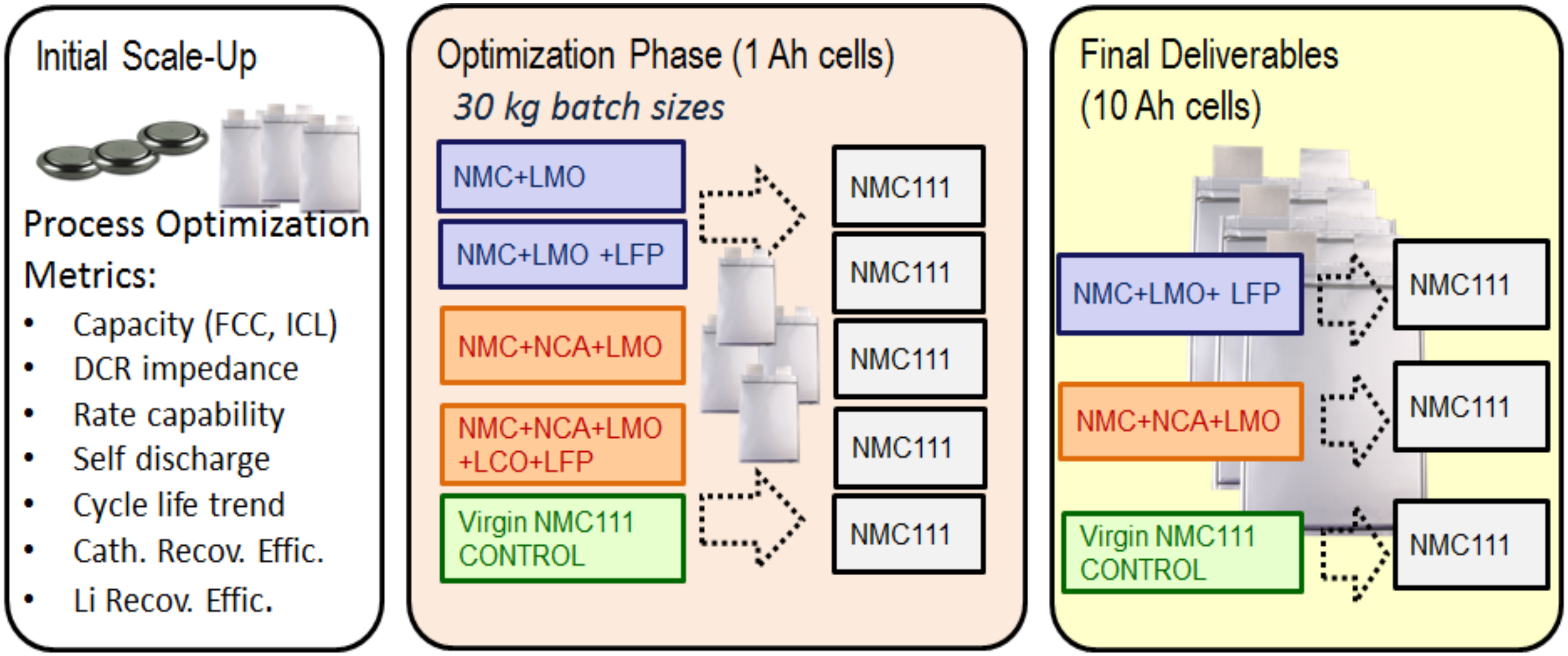
Relevance and Project Objectives

- Recycle multiple 10 kg size batches of end of life EV batteries consisting of different incoming cathode chemistries
- Produce cells of a single chemistry that could be used in a PHEV battery, to be tested according to USABC's PHEV test methods
- Improve the performance of the recovered cathode materials so that they exhibit performance on level with current commercial materials
- Recycle other materials including steel, copper, aluminum, etc.

Milestones

Tasks and Milestones	# of Cells	Q1, Yr1	Q2, Yr1	Q3, Yr1	Q4, Yr1	Q1, Yr2	Q2, Yr2	Q3, Yr2	Q4, Yr2
		Timeline (months)- 2 Year Duration							
Task 1:Improve electrochemical performance using 1 kg batches									
1.1 Optimize NMC111 synthesis parameters									
pH, material adding rate, temperature, etc are determined.									
Milestone 1.1: Recovered NMC111 shows similar properties to commercial NMC111									
1.2 Improve lithium and cathode material recovery rate									
Determine how much water needs to be evaporated.									
Optimize pH and concentration of impurity removal									
Milestone 1.2a: Li recovery efficiency >70%									
Milestone 1.2b: Cathode recovery efficiency >80%									
1.3 Electrodes Development									
Cathode Evaluation: Coin Cell, Full-Cell									
Milestone 1.3: Full cell testing shows properties within 5% of virgin material									
Deliverable: 100 coin cells to A123/WPI	100								
Task 2: Scale Process to 30 kg batch size									
2.1 Verify the state of health of spent batteries									
2.2 Scale process to 10 kg batch size									
Scale up the recycling process to 10kg from 1kg per experiment									
Determine the recycling efficiency									
Milestone 2.1: Scaled process cathode powder within 5% of control									
Milestone 2.2a: Li recovery efficiency >70%									
Milestone 2.2b: Cathode recovery efficiency >80%									
2.3 Scale process to 30 kg batch size									
Scale up the recycling process to 30kg from 10kg per experiment									
Determine the recycling efficiency									
Milestone 2.3: Scaled process cathode powder within 5% of control									
Milestone 2.4a: Li recovery efficiency >70%									
Milestone 2.4b: Cathode recovery efficiency >80%									
Milestone 2.5: Scaled process 1 Ah cells within 5% of control cells									
Task 3: Produce 200 cells from recycled material (4 input streams+control)									
Cell Assembly and Formation									
Performance Testing									
Cycle life									
Calendar Testing									
Milestone 3.1 Recovered cathode powder from each lot within 5% of control									
Milestone 3.2 Recovered cathode cells from each lot within 5% of control cells									
Deliverable: 200 1Ah cells to USABC/A123	200								
Task 4: Fabricate large format prismatic 10 Ah cells (A123)									
Milestone 4.1 Recovered cathode powder from each lot within 5% of control									
Milestone 4.2 Recovered cathode cells from each lot within 5% of control cells									
Deliverable: 60 10Ah cells to USABC/A123	60								
Final Report									

Cell Fabrication and Test Plan



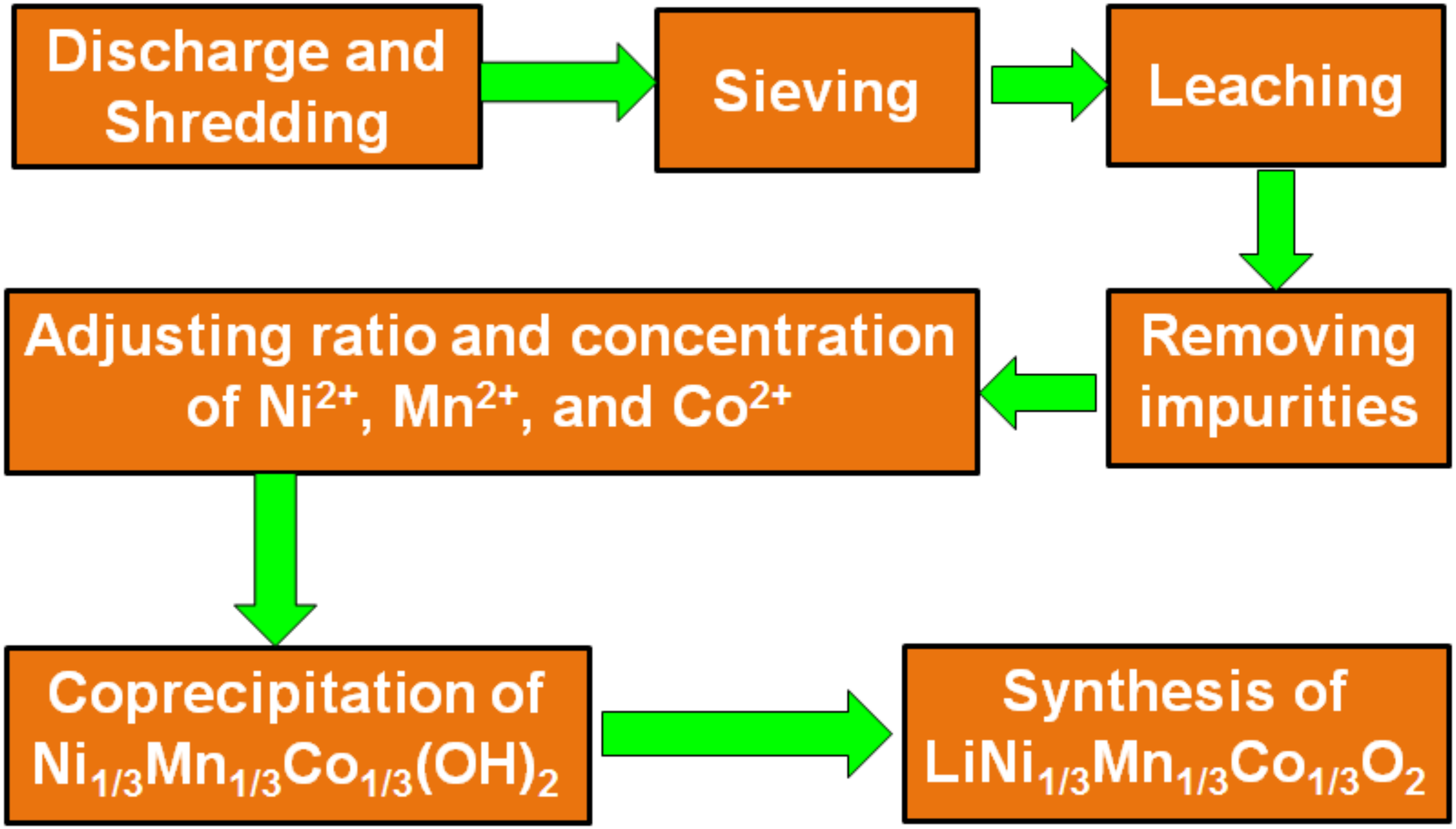
Test Article	Total Made	Groups	USABC/ ANL	A123/ WPI	Comments
1 Ah NMC111/ gr cells	250	4	(4x10) 50 total	200	Less cells have been fabricated due to the low usage of materials.
10 Ah NMC111/ gr cells	60	3	(3x10) 30 total	30	Less cells have been fabricated due to the low usage of materials.

Approach: a Close Loop Process

Advantages:

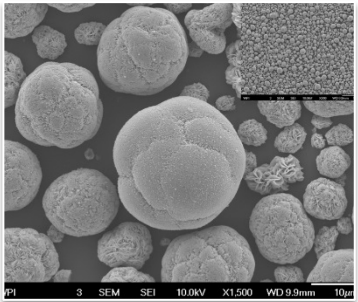
- Any lithium Ion battery
- Any size and shape
- No sorting
- Synthesize new $LiNi_xMn_yCo_zO_2$ directly
- Ratio of Ni, Mn and Co can be specially tailored to customer demands

Technical Accomplishment and Progress: Recycling Process Has Been Developed

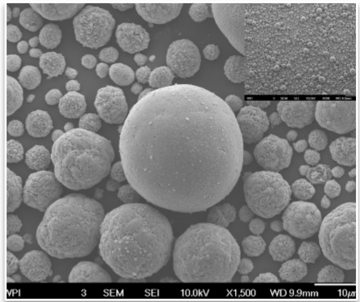


Recycling process is developed to offer a close loop process for the end of life EV batteries.

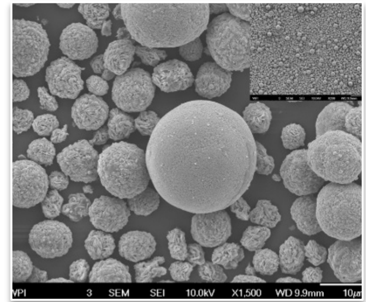
Morphology of Precursor (NMC Hydroxide)



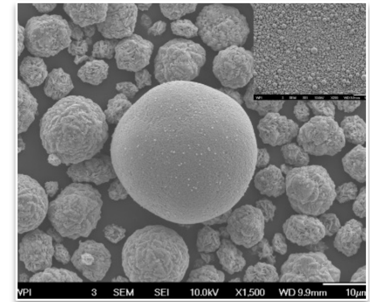
Day 1
1.91 g/mL



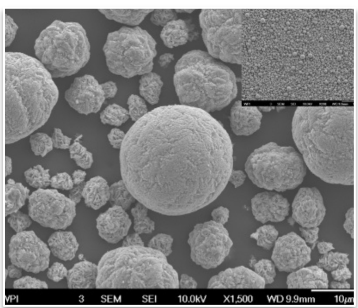
Day 2
1.99 g/mL



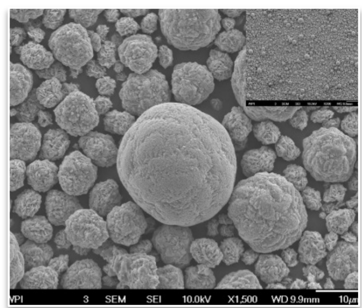
Day 3
1.98 g/mL



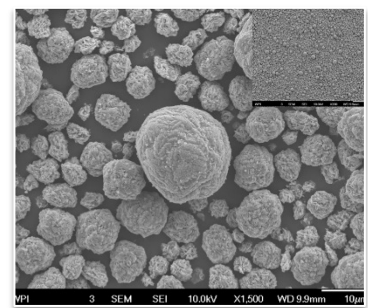
Day 4
2.02 g/mL



Day 5
1.99 g/mL



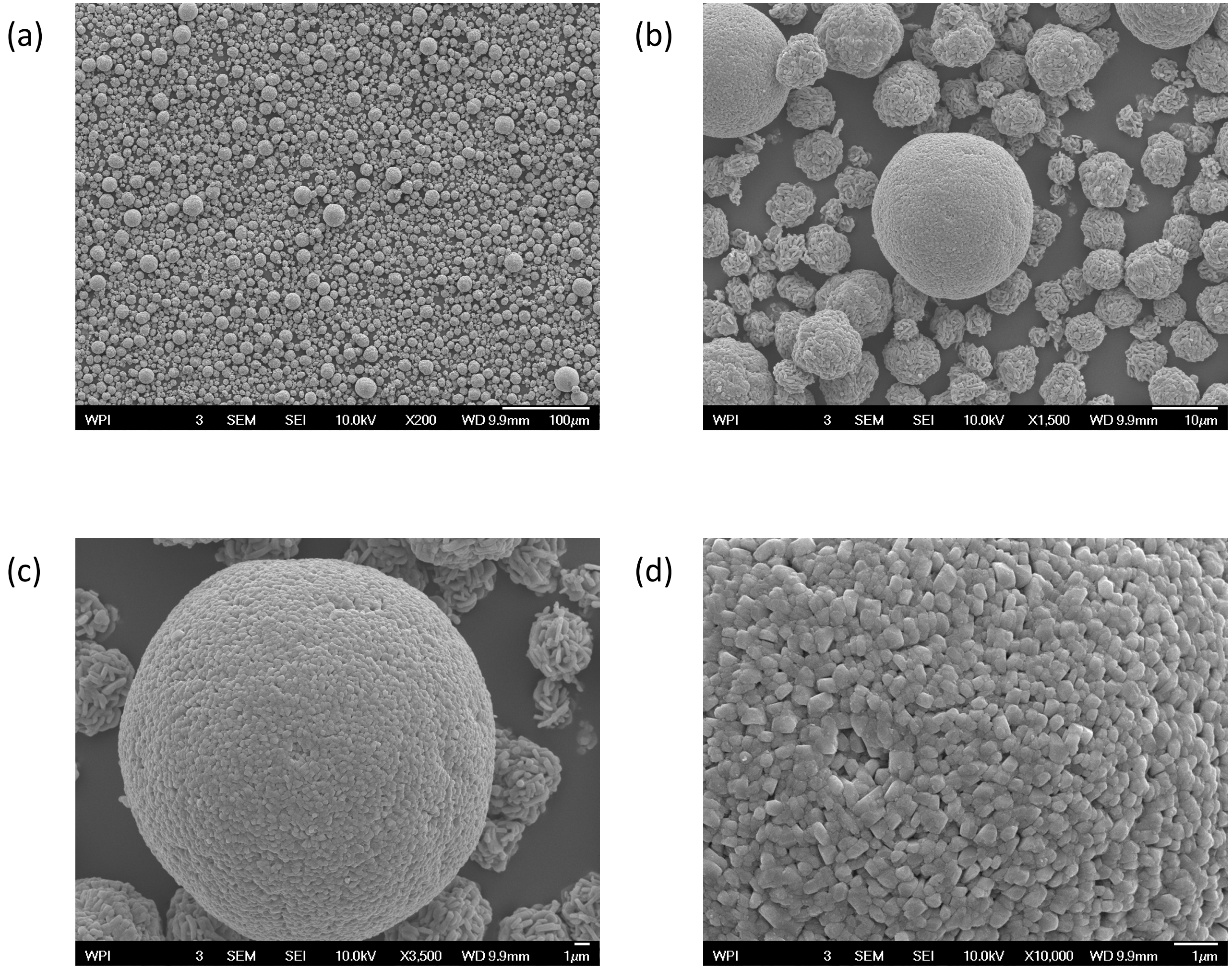
Day 6
1.97 g/mL



Day 7
1.96 g/mL

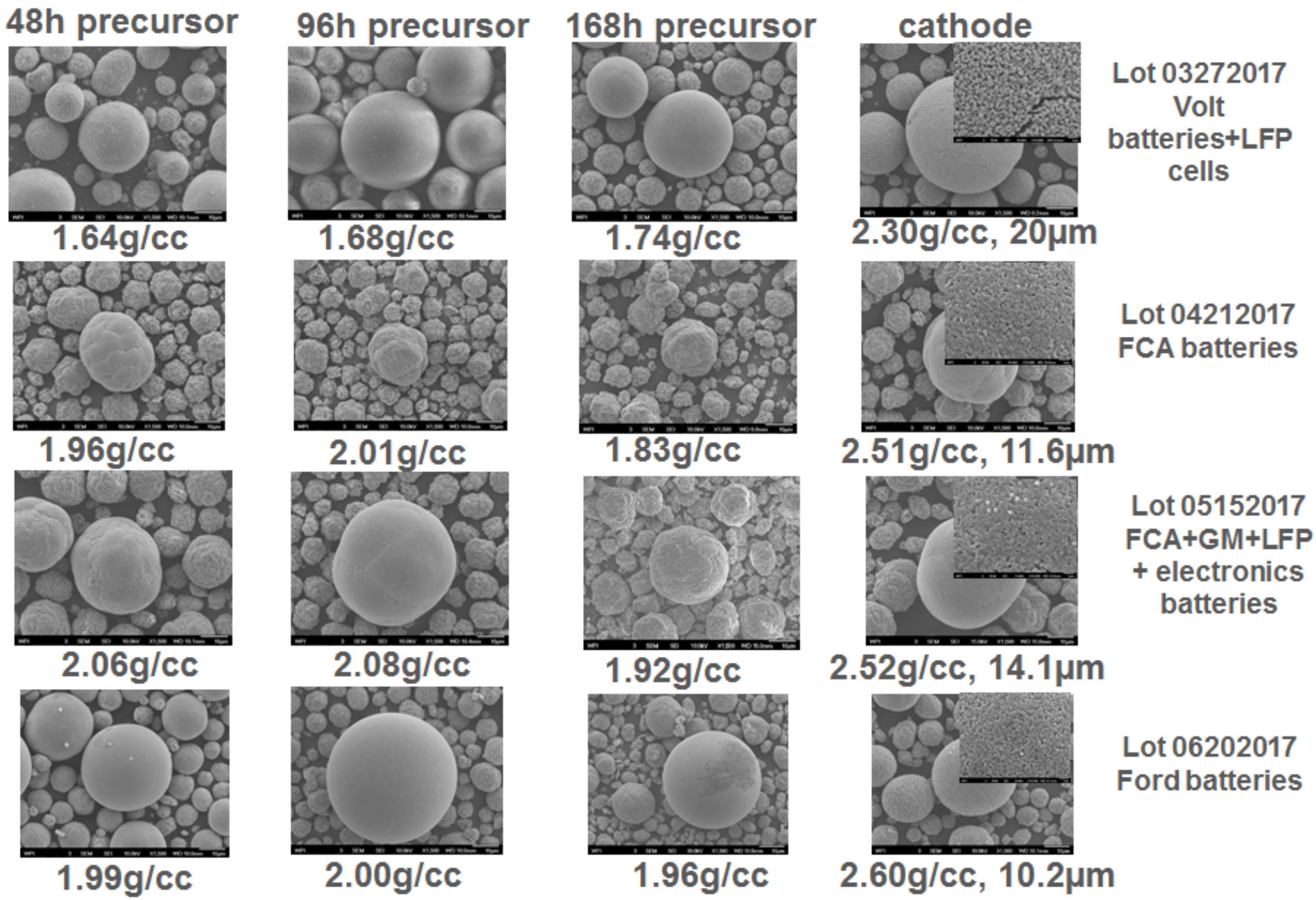
NMC precursor with good morphology has been synthesized with the recycled materials.

NMC111 Powder



Spherical and high tap density (>2.5g/cc) NMC111 powder is synthesized from the recycling stream.

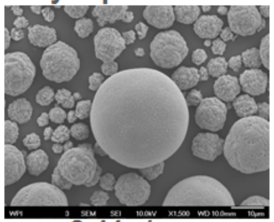
NMC111 from Different Recycling Streams (for 1Ah cells)

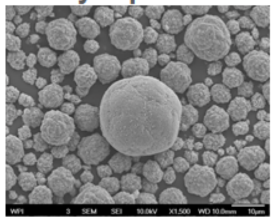


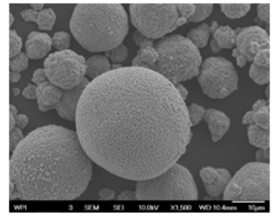
NMC111 powder has been synthesized from different recycling streams. The powder is used for 1Ah cells.

NMC111 from Different Recycling Streams (for 10Ah cells)

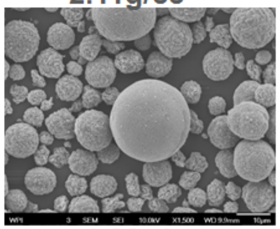
Day 3 precursor | **Day 7 precursor** | **Cathode**

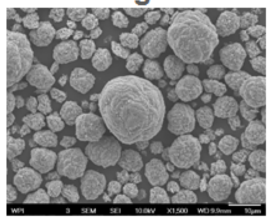

2.11g/cc

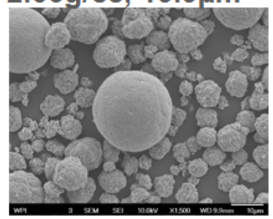

2.12g/cc


2.56g/cc, 13.9µm

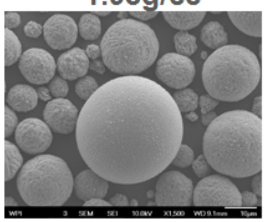
Lot 08112017
First experiment
Ford Focus batteries+LFP cells

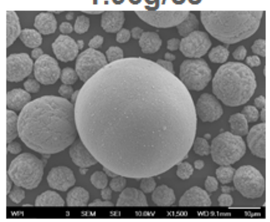

1.98g/cc

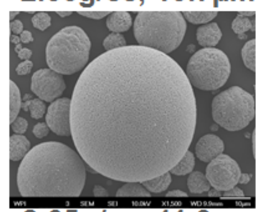

1.96g/cc


2.56g/cc, 10.7µm

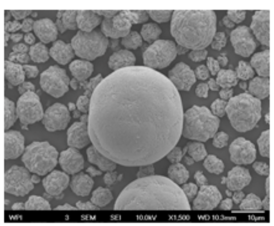
Lot 08302017
Second experiment
Ford Fucus batteries+LFP cells

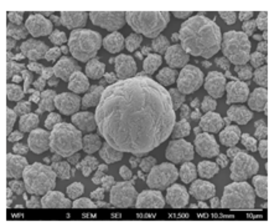

2.02g/cc

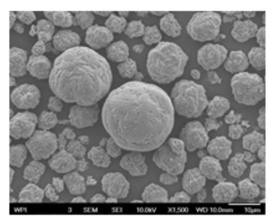

2.10g/cc


2.65g/cc, 11.9µm

Lot 09192017
Third experiment
FCA cells


2.05g/cc


2.03g/cc


2.50g/cc, 9.81µm

Lot 10092017
Fourth experiment
FCA cells

NMC111 powder has been synthesized from different recycling streams. The powder is used for 10Ah cells.

ICP Results-Impurity of NMC 111

Elements	Commercial NMC 111 Cathode (mol/L)	WPI NMC 111 Precursor (mol/L)	WPI NMC 111 Cathode (mol/L)
Ni	0.386	0.195	0.522
Mn	0.335	0.199	0.514
Co	0.365	0.199	0.515
Li	1.078	0.000	1.591
Na	0.014	0.000	0.000
Cu	0.000	0.000	0.000
Fe	0.000	0.000	0.000
Al	0.008	0.010	0.009
Mg	0.000	0.000	0.000

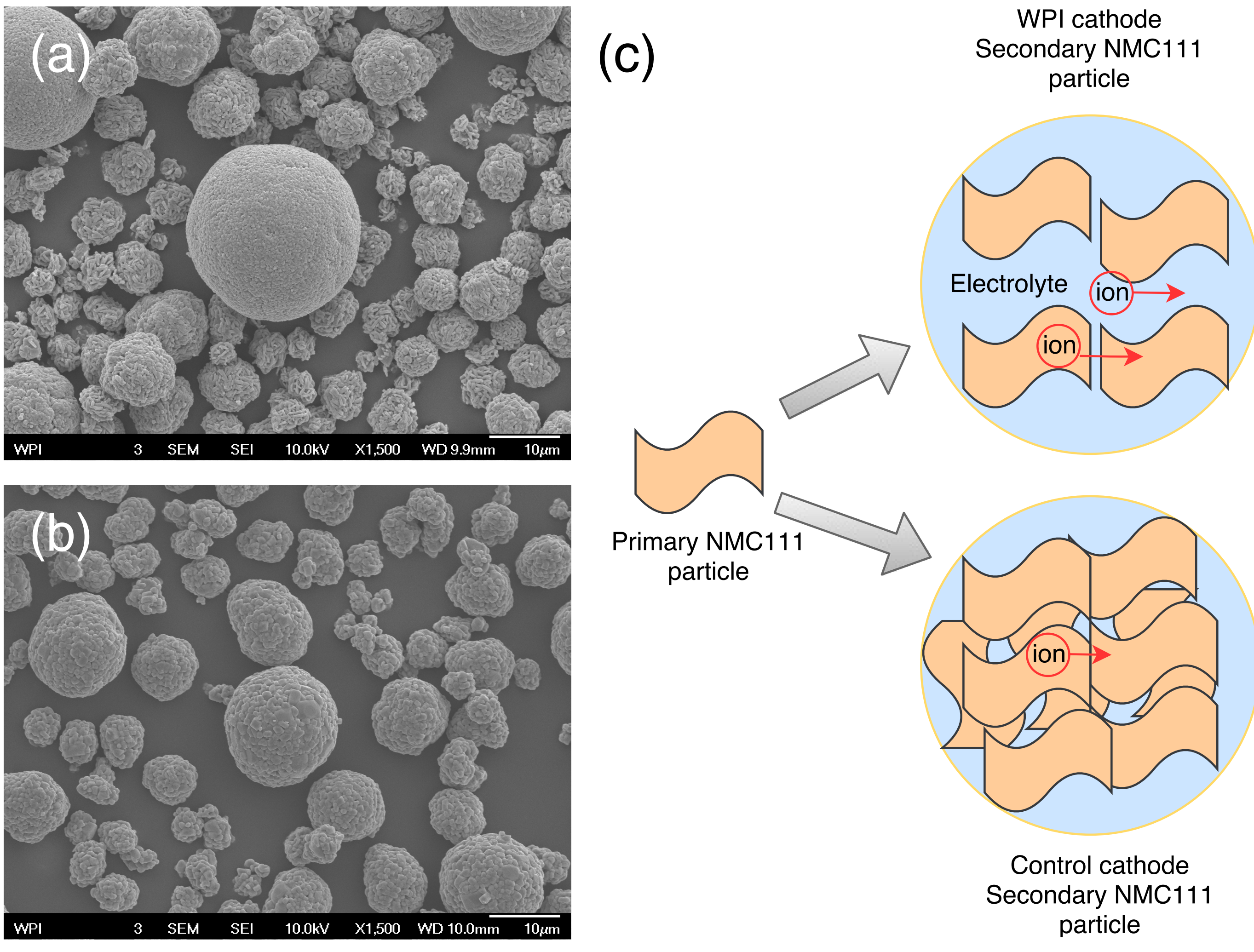
Recovered NMC111 precursor and powder have low impurity levels.

A123 Test Results (Coin Cells)

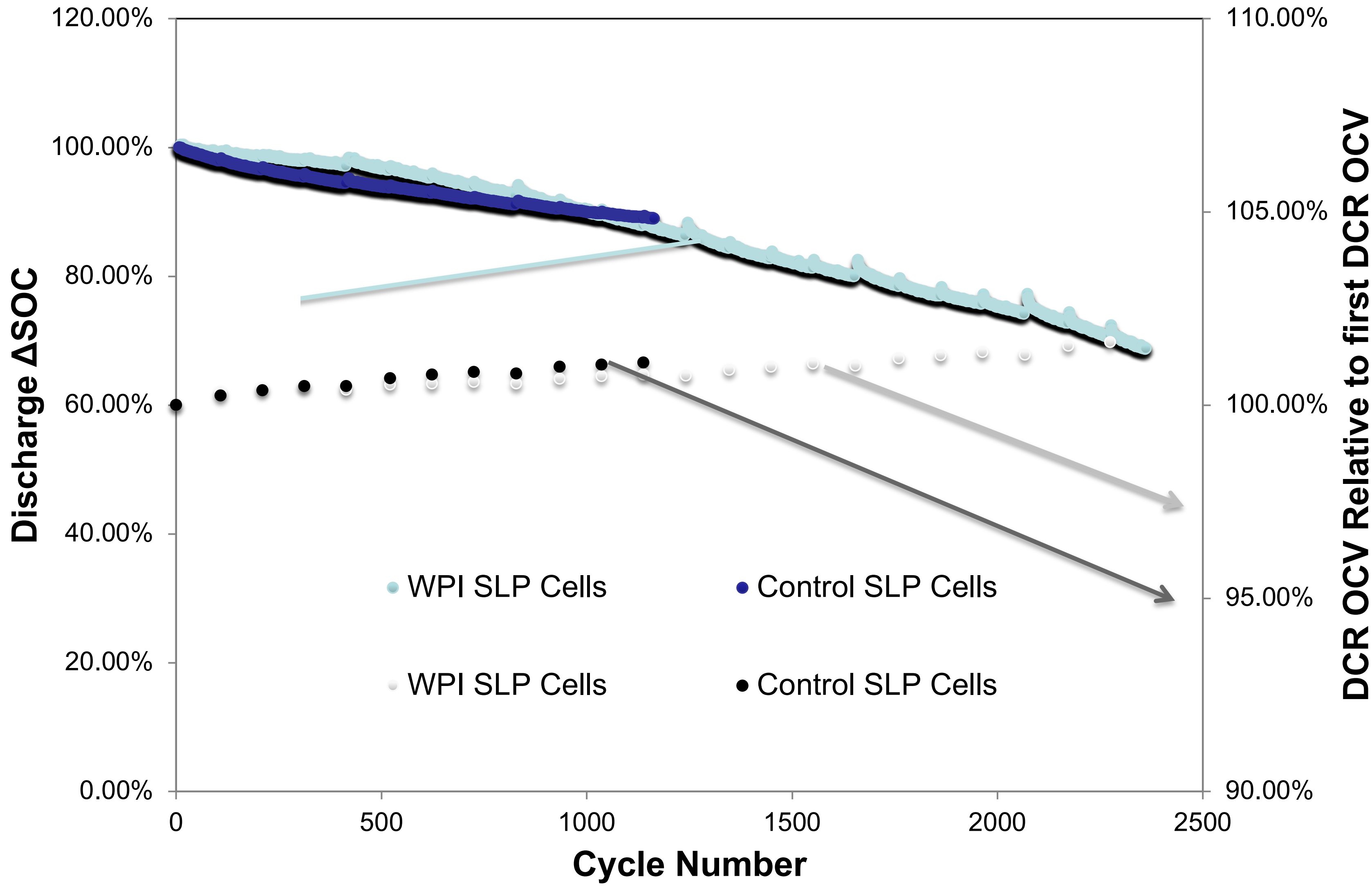
Test	Metric	Control Powder (B)	09112017 8 kg	10182017 8 kg
Tap Density	g/cc	2.84	2.45	2.52
D50 PSD	um	9.2	11.7	11.2
BET	m2/g	0.28	0.40	0.43
FCC/ FDC	mAh/g	174.7/ 157.2	168.9/ 148.6	179.6/ 156.1
Efficiency	%	90.0	88.0	86.9
1C	mAh/g	130.9	136.3	139.2
2C	mAh/g	119.4	130.0	131.6
5C	mAh/g	40.4	107.5	111.2
Full coating		Complete	Complete	Complete

Recovered NMC111 powder has comparable performance comparing to the commercial powder. The high rate performance of recovered powder is better.

Possible Reason of Rate Capability Difference



Single Layer Pouch Cell Performance



Cost Analysis

10 tons/ day recycled

Cathode Production			
	Pyro	Hydro*(BR)	Virgin
Cost	$41.63	**$31.40**	$39.09
Energy use in MJ per kg cathode material produced			
Total Energy	175.151	**143.765**	179.518
Fossil fuels	162.011	**134.362**	165.545
Coal	39.883	**29.720**	40.745
Natural gas	83.952	**70.093**	83.408
Petroleum	38.176	**34.549**	41.392

5 tons/ day recycled

Cathode Production			
	Pyro	Hydro*(BR)	Virgin
Cost	$39.50	**$33.71**	$39.09
Energy use in MJ per kg cathode material produced			
Total Energy	170.027	**147.783**	179.518
Fossil fuels	157.115	**138.380**	165.545
Coal	39.116	**29.720**	40.745
Natural gas	80.478	**70.093**	83.408
Petroleum	37.520	**34.549**	41.392

- Modelling BRs economic process with ReCell from ARL
  -BRs process modelled with the hydro model
  -Pyro is the cost to make cathode materials via smelting EV batteries
- Input was 50% LMO 50% NMC EV batteries
- Output was NMC111

Responses to Previous Year Reviewers' Comments

Comment 1: The reviewer replied that this is a very practical project and approach to recycling but asked how is this recycling process different/novel/more cost-effective than Umicore's approach. The reviewer also stated that much more electrochemical testing is required to conclude that the recycled materials are of adequate quality.

Response: Umicore is using a pyrometallurgical process to recycle lithium ion batteries. In their process, they burn all the batteries and only recover Co, Ni and Cu. In our recycling process, we directly synthesize high quality NMC cathode powder (cathode powder accounts for >50% of the material value in lithium ion batteries.). In addition, our recycling process allows us to recover most of the materials (>90wt%).

Comment 2: The reviewer had two comments: First, the activities listed as remaining challenges are too sparse, elaborating that here is not enough electrochemistry data to jump to 2 Ah cells. The reviewer said that much more testing will be needed and that this should be done before scaling up the process. Second, a full economic model should be included to answer questions such as what do the waste streams look like now and is the cost of managing those included here.

Response: We only listed part of the electrochemistry data due to the limited slide number. We have done detailed economical model to show the economical feasibility of our recycling process.

Comment 3: The reviewer stated that there is good analytical comparison, but a lack of electrochemical evaluation and asked if the team has done a cost assessment.

Response: Our collaborator (A123 Systems) has done very detailed electrochemical testing with coin cells, single layer pouch cells, 1Ah cells and 10Ah cells.

Collaboration and Partners

BATTERY RESOURCERS — Scale-up and commercialization

A123 SYSTEMS — Fabricate commercial cells

SNT SPIERS NEW TECHNOLOGIES — Disassemble EV battery packs

Argonne NATIONAL LABORATORY — Evaluate cells fabricated with recycled materials

Ford Go Further, FCA FIAT CHRYSLER AUTOMOBILES, GM — Provide battery packs

Remaining Challenges and Barriers

- Further Scale up to commercialization level

Proposed Future Work

- Recover other NMC materials with recycled streams
- Determine the impacts of different anode and adhesive materials
- Optimize the economical model

Summary

- The recycling technology offers a close loop process and can recycle Li-ion batteries with any shape, size and chemistry and sorting is not needed.
- The synthesized $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ from different recycling streams shows promising material and electrochemical properties.
- The process has been scaled up to 30kg spent batteries per experiment.
- Cost model shows the economical feasibility of the WPI recycling technology.